Transmutation Schemes for LWR's

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- **Transmutation with the objective to lower the burden on a repository as far as:**
  - *masses/volumes*
  - *heat load*
  - *doses/radiotoxicity*

has been studied both in Fast neutron systems and in thermal neutron systems.

- **Here we summarize findings related to transmutation in LWRs.**

Transmutation in LWRs

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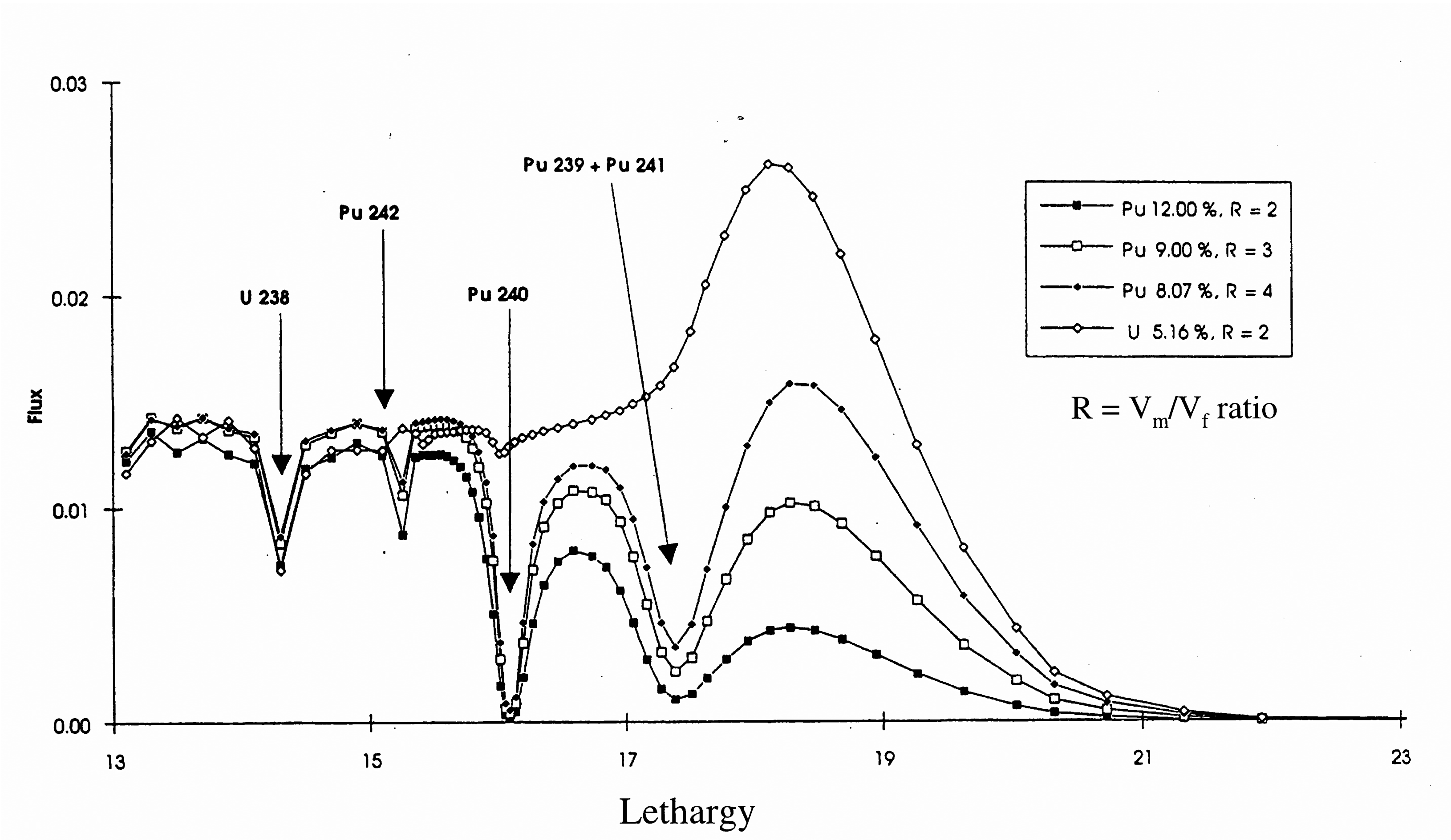
- **To reach the objective stated previously, one can use two approaches:**
  - Use of multirecycling. In this case the "key" parameter which defines the performance achievable with transmutation is the process **separation efficiency** for Pu and MA (e.g. 0.1% losses to the wastes)
  - Use of long irradiation time for a once-through-then-out (OTTO) approach. In this case the "key" parameter is the **"cumulative fission rate"** (e.g. 90% or more of fissions in the fuel/target)
- **As far as fuel forms, two techniques:**
  - Homogeneous recycling of Pu and MA
  - Targets (or dedicated, i.e. U-free, fuels)
- **For MA, also two strategies (Np goes always with Pu):**
  - Am only – Cm to the wastes or to intermediate storage
  - Am + Cm

"MULTIRECYCLING" Approach – Pu first!

- The first requirement is to adapt any MA strategy to the best option for an effective Pu recycling.
- There is in fact a basic difficulty in Pu multirecycling in PWRs, related to the degradation of the Pu vector, the subsequent need to increase the Pu content, with a hardening of the neutron spectrum, up to the point that the coolant void coefficient becomes positive due to the reduced absorption in the ˜ eV resonances of Pu-240 and Pu-242.
- Since the multirecycling of Pu induces a relevant increase of the MA production (with respect to the OTTO strategy with UOX), one has also to minimize the ratio R:

$$R = \frac{MA\ production}{Pu\ consumption}$$

- **From a physics point of view, the ratio R is closely related to cross-section ratio** $\alpha = \overline{\sigma}_c / \overline{\sigma}_f$ **of the TRU and its variation with the hardness of the spectrum.**
- **In order to vary the spectrum hardness, one can play with the V_m/V_f moderator-to-fuel ratio.**
- **To stay in a realistic range, the V_m/V_f ratio can be varied between 1.3 and 4 (standard PWR value : ~ 2). The studies performed show that the high V_m/V_f ratio (~ 4) should be chosen, in order to minimize the ratio R.**
- **For example, for $V_m/V_f = 2$ R = 0.22**
  $V_m/V_f = 4$ R = 0.12


0.03
0.02
0.01
0.00
Flux
U 238
Pu 242
Pu 240
Pu 239 + Pu 241
Pu 12.00 %, R = 2
Pu 9.00 %, R = 3
Pu 8.07 %, R = 4
U 5.16 %, R = 2
R = V_m/V_f ratio
13
15
17
19
21
23
Lethargy

- **However due to the more thermalized spectrum, the degradation of the Pu vector is very significant. This is an example:**

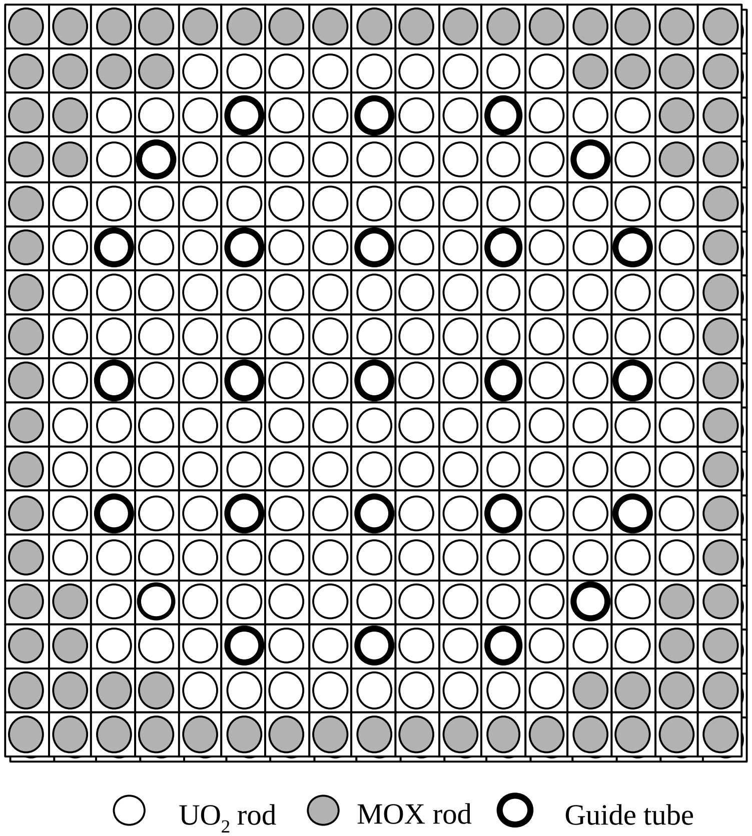
	End of cycle		Initial
	PWR-MOX $V_m/V_f = 2$	PWR-MOX $V_m/V_f = 4$	
Fissile Pu/Pu	51.5%	35.8%	64.1%
238 Pu/Pu	3.7	3.9	2.7
239 Pu/Pu	37.9	21.3	56.0
240 Pu/Pu	30.7	36.7	25.9
241 Pu/Pu	13.6	14.5	8.1
242 Pu/Pu	10.0	20.3	7.3

- **This has as a consequence that, even with dilution with better quality Pu, after 2÷3 recyclings, a positive coolant void coefficient is obtained. Also, after a few recyclings, due to the high production of Pu-242, the ratio R would become worse or less favorable.**

- The best option which has been found is to multirecycle the Pu on an enriched-^{235}U support:
  - *The MIX concept (homogeneous mixing of U and Pu)*
  - *The CORAIL concept (heterogeneous assembly)*
- Both concepts look feasible and they provide a mean to stabilize the Pu stocks, with a progressive introduction of these new PWRs to replace UOX-PWRs over several decades.

  No major drawbacks are found, both on the reactor parameters and on the fuel cycle characteristics. However, potential power peaks should be optimized. Significant fuel and assembly design validation would be needed, in order to cope with licensing requirements.
- The addition of Np to Pu has not been explicitly envisaged, but it should not change dramatically nor the core parameters nor the fuel cycle characteristics.

CORAIL Multirecycle Concept for Deep Burnup in Thermal Systems



French-CEA CORAIL concept considered for Pu or TRU stabilization (i.e., no net production of Pu or TRU)

Compatible with existing LWR

- **Concept**
  - Heterogeneous assembly in a homogeneous core
    - *Mitigates adverse effect on reactor control parameters*
  - Standard design using fuel rods and assembly that are qualified
    - *Mass balance in CORAIL core is similar to 30% MOX case, but much better for multirecycling*
  - Pu/TRU discharged from both MOX and UOX pins is recycled
- **Design Criteria**
  - Uranium enrichment < 5.0% (enriched uranium supports mission)
  - Pu content in MOX < 12%
  - Power peaking factor < 1.2

“MULTIRECYCLING” Approach.
The addition of MA

- **Some physics facts:**
  - In a PWR spectrum, most TRU have unfavorable $\alpha = \bar{\sigma}_c / \bar{\sigma}_f$ values. As indicated previously there are major consequences:
    - *Tight neutron balance, due to positive (neutron consumption)/fission (D parameters) for most TRU*
    - *Degradation of the Pu vector under irradiation*
    - *High production of higher mass MA*
  - The addition of MA to Pu has generally unfavorable effects, as summarized in the following table:

Spectrum Flux ($n/cm^2/s$)	FR 10^{15}	PWR-UOX 10^{14}
^{234}U	-0.42	+0.35
^{235}U	-0.86	-0.60
^{236}U	+0.14	+1.94
^{238}U	-0.62	+0.10
^{237}Np	-0.55	+1.10
^{238}Pu	-1.33	+0.15
^{239}Pu	-1.46	-0.69
^{240}Pu	-0.91	+0.42
^{241}Pu	-1.21	-0.57
^{242}Pu	-0.49	+1.27
^{241}Am	-0.54	+1.07
^{242m}Am	-1.87	-1.54
^{243}Am	-0.67	+0.32
^{243}Cm	-2.19	-1.89
^{244}Cm	-1.47	-0.66
^{245}Cm	-2.68	-2.29
^{246}Cm	-2.15	+0.08
^{247}Cm	-2.46	-0.93
^{248}Cm	-2.26	-0.23
Pu as unloaded from PWR	-1.23	-0.25
MA as unloaded from PWR	-0.61	+0.91
TRU as unloaded from PWR	-1.17	-0.14

Parameter D: (neutron consumption)/fission

Addition of	Neutron Balance. D_{fuel}	Core performances. Void coefficient	Fuel cycle Doses at fabrication
Np	D_{fuel} decrease	Less margin to positive	Slight dose increase
Np+Am	D_{fuel} decrease	Idem	Significant increase
Np+Am+Cm	D_{fuel} up (with respect to Np+Am)	Slight improvement (with respect to Np+Am)	Neutron source dramatically increased

- Consequences on reactor parameters impose limitation of the maximum percentage of MA allowed in the fuel (typically ~1% if $V_m/V_f = 2$; ~2% if $V_m/V_f = 4$).
- The neutron balance constraint impose an increase in U enrichment/Pu content.
- Fuel cycle consequences are a major issue and can make multirecycling not practical.
- The cases of MA in CORAIL and MIX have been worked out in a rather detailed manner. For CORAIL, results obtained at ANL and CEA agree fairly well. Agreement also on the conclusions.

Radiation Properties (per 1 MT-HM) with Various Recycle Hypotheses

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Recycle Elements		UO2	Pu	Pu-Np	Pu-Np-Am	TRU
Decay Heat (kW)	Charge	0.00001	0.7	1.2	3.8	11.9
	Discharge	2059	2002	2008	1993	1999
	After cooling	2.5	4.4	4.9	9.6	14.5
Neutrons (n/sec)	Charge	1.23E+04	3.96E+07	6.05E+07	1.50E+08	1.51E+12
	Discharge	1.23E+09	9.59E+09	9.45E+09	2.50E+10	9.08E+12
	After cooling	5.74E+08	6.36E+09	6.00E+09	1.51E+10	2.51E+12
Gamma Source (kW)	Charge	0.	0.0003	0.0005	0.003	0.007
	Discharge	567	548	549	538	534
	After cooling	1.07	1.03	1.02	0.9	1.0

- **Decay heat is considerable higher when Am is recycled**
  - Resulting heat loads comparable to cooled spent fuel
- **Neutron sources much higher with TRU recycle**
  - Generated by Cm-244, and Cf-252 with multi-recycle
- **Gamma source is much lower in heavy metal than spent fuel where the fission products dominate**

MULTIRECYCLING:
Targets for heterogeneous recycling?

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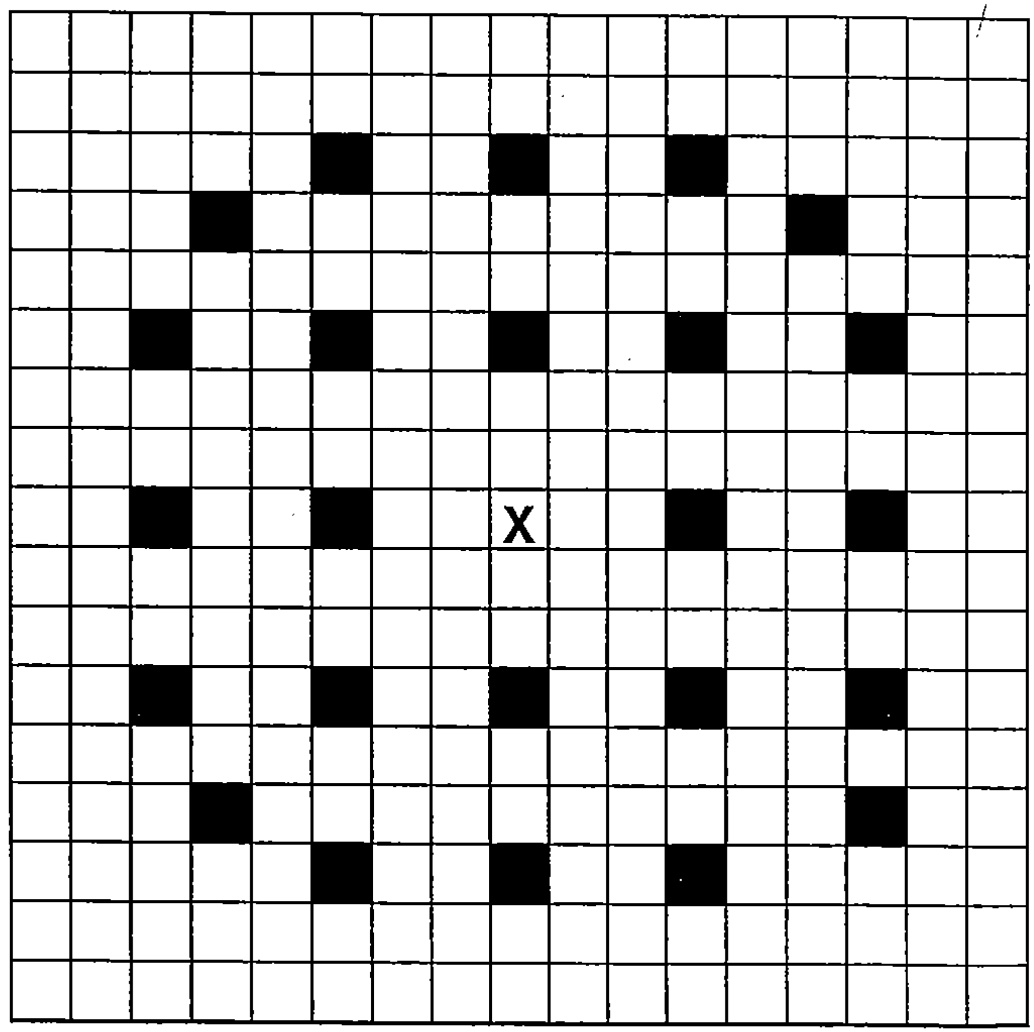
- **To avoid to "pollute" the whole fuel cycle (e.g. fabrication) with MA, one can think to concentrate MA in targets, to be put in specific PWR assemblies, and allow for a specific MA target management.**
- **A concept has been studied in detail at CEA: the TIGRE concept.**
  - MA targets (Am or Am+Cm) are put in the guide tubes of standard PWR assemblies.
  - The MA content (on an inert support) is optimized to avoid power peaks during irradiation, to avoid too long irradiation times and still allow a significant transmutation rate for each PWR equipped with "TIGRE" assemblies.

- For targets with Am only, an equilibrium (i.e. productions=destruction in the reactor) can be reached, but almost all PWRs have to be equipped with TIGRE assemblies (~70 for each PWR). This number is imposed to limit the overenrichment in ^{235}U (PWR-UOX) or Pu content (PWR-MOX).
- The linear power in the target at the end of irradiation is 130 W/cm, which can be a problem for cooling, according to the design of the guide-tubes in different PWRs.
- If (Am+Cm) is put in the targets, the number of TIGRE assemblies required increases beyond the constraint of allowable assemblies (constraint on enrichment) and an equilibrium cannot be found.
- Limited gains on the parameters of a repository are obtained with the “Am – only” strategy. (E.g. a further reduction of a factor of 2÷3 on radiotoxicity with respect to the “Pu-only” recycling scenario.)



PWR-UOX assembly with MA targets

? Standard UO_2 pin

¦ Guide tube with targets

X Instrumentation tube

24 targets for each assembly

Each target: 30% AmO_2 + 70% Al_2O_3

MULTIRECYCLING Approach (in LWRs): Conclusions

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- **Best is homogeneous recycling**
- **Best when done in MOX-PWR with ^{235}U support (MIX or CORAIL), despite the fact that $V_m/V_f = 2$ is not optimum**
- **Consequences on the fuel cycle (e.g. increase of doses and neutron sources) can prevent to implement full MA multirecycling (some difficulties on local power peaking also).**
- **The Am-only multirecycling has modest impact on masses/doses reduction. Moreover, a satisfactory strategy for separated Cm has still to be found and can be rather hard to find.**

THE "CUMULATIVE FISSION RATE" Approach

This approach has been implemented in two different ways:

- **Heterogeneous monorecycling of MA targets (TIGRE and ANDIAMO concepts)**
- **Fertile-free fuels (Pu+MA), to increase the transmutation potential. This approach has been applied to:**
  - Full core with (Pu+MA) – fertile free fuel
  - Segregating (Pu+MA) fuel in selected pins in a standard PWR-UOX assembly (A "hybrid" approach: the APA concept, with fertile-free pins and multirecycling)

"The CUMULATIVE FISSION APPROACH": Heterogeneous Mode

- **The monorecycling of targets in the TIGRE assembly has been attempted, both for "Am-only" and for Am+Cm targets.**
- **The major results are as follows ("Am-only" targets):**
  - For each PWR-UOX, 66 assemblies with 24 targets are introduced.
  - To reach 90% accumulative fission rate, 25 years irradiation are needed. To reach 97%, 40 years are needed.
  - Since to reach these high fission rates one has to decrease the amount of Am in each target, the transmuted masses are modest (2.5 to 1.5 Kg/TWhe). To increase that, there is a need to overenrich in ^{235}U.

    In any case, the power variation during irradiation is very significant.

- **An alternative approach: a "dedicated" assembly (ANDIAMO), with MA-based pins on a fissile support.**
- **The loading of the targets in heavy atoms is 60% by mass in an inert matrix. The composition is: ~50% Pu, 50% MA.**
  - The power and reactivity variations are optimized and the "dedicated" assembly can be made "transparent" with respect to the surrounding PWR assembly environment.
  - One can reach a sizable destruction rate (~5.8 Kg/TWhe of Am), which allows to get equilibrium in a reactor park.
- **However, irradiation times are still too long to be realistic (~30÷40 years to reach 96% fissions).**

Conclusions on the "CUMULATIVE FISSION RATE" Approach

- **This approach has practical limits (difficult to reach >90% fission rates). Material problems can make the approach not feasible.**
- **The approach has been applied in France to targets (heterogeneous mode).**
  - No attempt to design an U-free fuelled core for once-through (examples in Japan, Switzerland)
  - For fertile-free fuels, back to multirecycling (the APA concept)

A Hybrid approach: Fertile-free fuel and multirecycling

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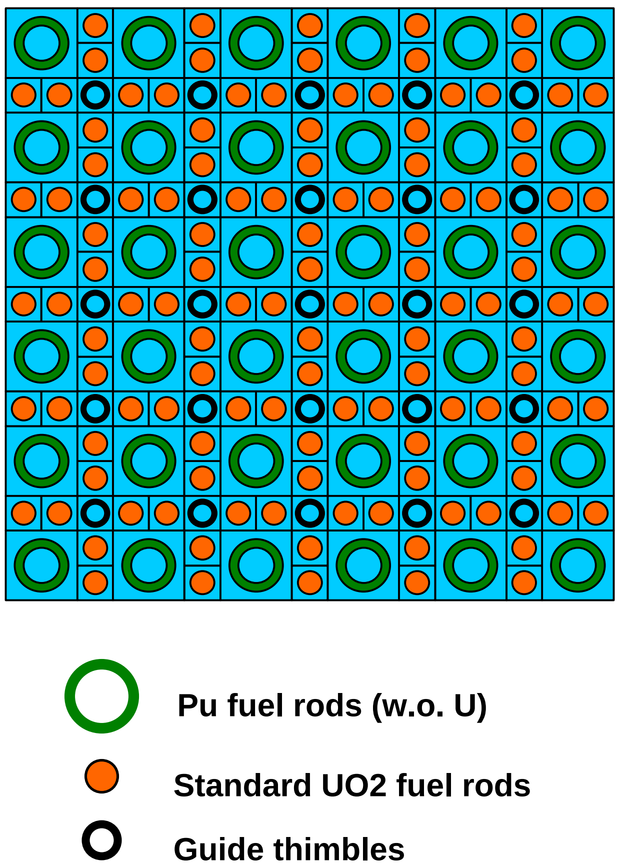
- The APA concept: A PWR assembly which uses 120 standard UO_2 rods and 36 large annular rods using Pu (or Pu+MA) fuel in an inert matrix (e.g. CeO2), with a high local V_m/V_f (~3.3).
- As for the CORAIL concept, in principle one can multirecycle Pu and MA up to stabilization of both Pu and MA inventories (in the French scenario, to ~260 tons of Pu and ~75 tons MA). The value for Pu is lower of what obtained with CORAIL.
- Even if not worked-out, the consequences on the fuel cycle of MA recycling are expected to be significant, but limited to only 10÷15% of the overall fuel fabrication.
- A similar concept is explored at MIT (CONFU concept).

Advanced Plutonium Assembly (APA) for Pu Multi-Recycling

APA subassembly



- **Concept**
  - Heterogeneous assembly in a heterogeneous core
  - Employs large fertile-free fuel [(Pu,MA,Ce)O2] pins in a typical 17x17 PWR assembly design
  - ~35% of APA in a typical core
  - Allows Pu stabilization at low inventory

- **A systematic approach to U-free fuels in PWRs has not yet been attempted (e.g. based on spectrum characteristics etc.) and should be done.**
- **The feasibility of most target or U-free concepts is related to fuel forms and performances to be experimentally validated (some experimental work underway on the fuels for the APA concept and in the frame of the PSI-Switzerland studies).**
- **Multirecycling should however be associated to U-free fuels to impact significantly on a repository. This will impose further requirements to the fuel form/matrix.**

CONCLUSIONS

- **Pu management in LWR's is feasible. Contrary to a past perception, multirecycling is feasible, and a few concepts seem to be acceptable (MIX, CORAIL …) within standard PWR's layouts.**
- **The main consequences of Pu multirecycling is the increase of MA build-up. This fact has consequences on the fuel cycle and potentially on the waste form (e.g. glasses).**

- **From a physics point of view, transmutation of MA in LWR's can be performed.**
- **However, several "prices" have to be paid:**
  - U-235 overenrichment or increased Pu content (tight neutron balance)
  - Severe consequences on the fuel cycle (e.g. at fuel fabrication) are expected for any option envisaged (high capture-to-fission ratios)
  - Careful re-examination of power distributions and reactivity coefficients (high sensitivity to neutron spectrum changes) are needed.
- **Only homogeneous recycling of <u>all</u> MA (including Cm), does effect significantly parameters like radiotoxicity.**

- **U-free fuels offer some potential advantage:**
  - Stabilization of Pu inventory to a lower level (increased Pu reduction)
  - Limitation to a part only of the fuel fabrication of the problems related to the MA recycling.
- **However, multirecycling should be envisaged also for this type of fuels.**
- **Fuel form development and validation: a major issue.**